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TRITIUM CONCENTRATIONS IN FOOD 1967-1968

A.A. Moghissi, et al

Southwestern Radiological Health Laboratory
Las Vegas, Nevada

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Tritium Concentrations in Food, 1967-1968

A. A. Moghissi and R. J. Lyon[1]

Tritium concentrations were determined in food samples obtained from several children's homes in the United States. The average concentrations of tritium in food indicate a decreasing trend from 1967 to 1968. The average dose equivalent from tritium in food was calculated to be 0.08 mrem for 1967 and 0.03 mrem for 1968.

Due to the biological significance of tritium, the Bureau of Radiological Health has for several years evaluated the radiological health aspects of this radionuclide. The results of studies dealing with surface waters, (*1*), rain (*2*), and urine (*3*) have been reported.

According to the recommendations of the International Commission on Radiological Protection (ICRP) (*4*) more than half of the tritium intake is through food. Uncertainties regarding the biological half life of tritium incorporated in the organic molecules of food justified an evaluation of the tritium present in food.

Sampling and analysis procedures

Food samples collected from several children's homes distributed throughout the continental United States were shipped to the Southeastern Radiological Health Laboratory. These homes were instructed to collect the total food intake of the children, including soft drinks, during a period of 24 hours. The only medium not included was tap water.

In the laboratory, the sample was homogenized and approximately 100–150 grams were combusted. The collected water was distilled and counted for tritium.

Combustion procedure

The details of the combustion technique are shown in figure 1. Air is passed through a column (D_1) containing a desiccant and molecular sieve (Linde 13 A) to remove water and organic compounds originating from air compressors. The purified air was passed through a copper oxide catalyst which was kept at 500 to 600° C. in a furnace (F_1). This step was necessary to oxidize hydrogen and methane in the air which are known to contain tritium (*5, 6*) at levels several orders of magnitude higher than atmospheric moisture. The resulting water was collected in the drying column (D_2). The sample (S) which was placed in the furnace (F_2) was partially combusted as the air passed by. The completion of combustion occurred in the furnace (F_3) in the presence of a copper oxide catalyst at 500–600° C. At the beginning of the process F_2 was gradually heated from room temperature to 600° C. within 2 hours. An additional 2 hours were necessary to remove the possible residues of the sample, making it possible to run two samples in a working day.

Subsequent to the completion of this work, the catalyst was replaced by a platinum-aluminum oxide catalyst[2]. This catalyst could be used at a lower temperature of 150–200° C. and thus was considerably more effective.

Accuracy of the combustion

Significant errors may be introduced in the results of chemical reactions dealing with tritium if the reaction is incomplete. This phenomenon is caused by isotope effect and is the basis for enrichment of deuterium and tritium.

To evaluate the reproducibility of this combustion system, approximately 5 g of a rat tissue from a tritium experiment was mixed with the food and combusted. The entire procedure was

[1] A. A. Moghissi is with Southwestern Radiological Health Laboratory, Las Vegas, Nev., and R. J. Lyon is with Southeastern Radiological Health Laboratory, Montgomery, Ala.

[2] The catalyst was obtained from the Englehard Industries, Newark, N.J.

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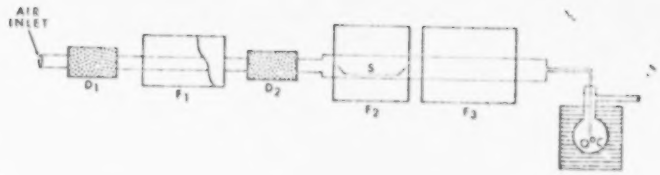

Figure 1. Schematic diagram of the combustion system

then repeated several times. The resulting water samples were distilled and counted as described below. Results of the replicates are shown in table 1. It can be seen that the standard deviation of the mean compares favorably with the counting error of the individual samples at the 2 σ confidence level.

Table 1. Accuracy of the food tritium analysis

Run number	Concentration in water of combustion (nCi/liter)
1	6.1 ± 0.3
2	5.9 ± .3
3	6.3 ± .3
4	6.0 ± .3
5	5.7 ± .3
Average	6.0 ± 0.2

Liquid scintillation counting

All samples were counted in a Beckman liquid scintillation counter. The details of the sample preparation are described by Moghissi et al (*7*). In short, 1 liter of scintillation liquid was prepared by dissolving 7 g of 2, 5-diphenyloxazole (PPO), 1.5 g of p-bis-(methystyryl)-benzene (bis-MSB) and 120 g of naphthalene in dioxane. Duplicate samples were prepared using 20 ml of this scintillation liquid and 5 ml of water. The total counting time for the duplicate was 200 minutes divided into four 50-minute counts. If any of the 50-minute counts deviated by more than 2 σ from the average, the duplicates were recounted. This occurred with a frequency comparable to the statistical expectancy. A background sample was counted prior to the first sample and a second background subsequent to its duplicate. The minimum limit of detection (MLD) of the counting was 0.2 nCi/liter at the 2 σ confidence level. Starting July 1968, one of the replicates was prepared using an emulsion mixture (*8*). A p-xylene solution containing 7 g PPO and 1.5 g bis-MSB per liter was mixed with Triton N 101 in a volume ratio of 2.75:1. Fifteen ml of this mixture were mixed with 10 ml of water resulting in a MLD of 0.1 nCi/liter. As this mixture was used in an experimental stage and for the sake of uniformity, all results are reported with an MLD of 0.2 nCi/liter and the counting error of the dioxane-based solution shown in figure 2.

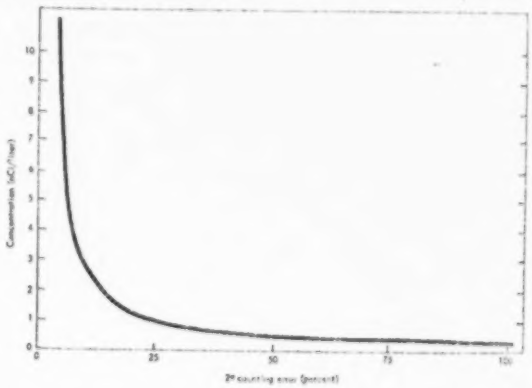

Figure 2. Correlation between the tritium concentration of water and the counting error in percent at 2-sigma confidence level

Results

The location of the stations and the analytical results are shown in tables 2 and 3. The decreasing trend as observed in surface water (*1*), rain (*2*) and urine (*3*) is evident in average values of 1968 as compared to 1967. A clear trend in individual samples is, however, not evident. The October 1968 value from Tampa, Fla., is considerably higher than other values during the entire period of 1967–1968. Similar values may be observed for other sample collection sites (Albuquerque, N. Mex., June 1967; Charleston, S.C., December 1967; etc.). This apparent lack of systematic decrease may be explained by the complexity of the food. Individual components of the samples could have originated from geographical areas

Table 2. Tritium concentration in food, 1967

Location	Concentration (nCi/liter of water of combustion)											
	January	February	March	April	May	June	July	August	September	October	November	December
Fla: Tampa	0.3	0.3	0.9	0.4	NS	NS	0.5	< 0.2	< 0.2	0.6	< 0.2	< 0.2
La: New Orleans	NS	.4	.5	.3	0.2	1.0	.6	1.0	< 0.2	.9	NS	1.0
Ark: Little Rock	< .2	.5	.4	.5	.2	.4	.3	< .2	NS	.3	.2	.4
Ky: Louisville	NS	NS	1.0	1.1	> .2	.8	.6	.8	NS	.6	.4	1.0
N. Mex: Albuquerque	.3	.6	.5	> .2	> .2	1.2	.5	NS	.3	NS	.6	NS
S. C: Charleston	NS	.6	.6	.3	1.5	> .2	> .2	1.1	.2	.4	NS	2.0
Miss: Columbia	.3	.3	.8	.4	> .2	1.0	.2	NS	.4	NS	< .2	< .2
Tex: Austin	.3	.6	1.0	> .2	> .2	1.0	NS	< .2	.5	< .2	< .2	< .2

NS, no sample received.

Table 3. Tritium concentration in food, 1968

Location	Concentration (nCi/liter of water of combustion)											
	January	February	March	April	May	June	July	August	September	October	November	December
Fla: Tampa	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	1.3	< 0.2	< 0.4
La: New Orleans	< .2	< .2	< .2	< .2	< .2	< .2	NS	< .2	< .2	.5	< .2	< .2
Ark: Little Rock	< .2	.3	< .2	< .2	< .2	< .2	< .2	< .2	< .2	< .2	< .2	< .2
Ky: Louisville	.4	.3	.3	.3	< .2	< .2	< .2	.3	.3	.3	NS	.2
N. Mex: Albuquerque	.5	.3	.4	.2	< .2	< .2	NS	.3	< .2	< .2	NS	.4
S. C: Charleston	.5	.3	.3	.3	< .2	< .2	< .2	< .2	NS	.3	.3	.5
Miss: Columbia	< .2	< .2	< .2	< .2	< .2	< .2	NS	< .2	< .2	.5	NS	.5
Tex: Austin	< .2	< .2	< .2	.2	< .2	< .2	NS	.2	.3	.5	NS	.6

NS, no sample received.

with different tritium concentrations than the collection site. Similarly, the preservation of food makes it possible that part of the food could have originated at a time when the tritium concentration was higher than that at the collection time. In the averaging process these effects are minimized so that a decrease is evident.

Dose calculation

In order that the dose equivalent to the population could be calculated, the average annual tritium concentration was used. For the purpose of averaging 0.1 nCi/liter was assumed to be the tritium concentration of values below the MLD. The following equation (*1*) was used for dose calculation:

$$DE = 10^{-3}(QF)C \qquad \ldots(1)$$

where

DE = dose equivalent in mrem/a;
QF = quality factor for tritium; and,
C = concentration of tritium in the body in nCi/liter.

Although ICRP has recommended a reduction of QF for tritium from 1.7 to 1, for the sake of uniformity with the dose calculations made for the same period using other media and for reasons described elsewhere (*3*), a QF of 1.7 is used in the dose estimations.

For the purpose of calculation it is assumed that the tritium concentration of the body is in equilibrium with that in food. It can be seen in table 4 that the average DE as calculated for food is somewhat lower than the averages calculated from body burden measurements for standard man of 0.15 mrem for 1967 and 0.13 mrem for 1968. This may be explained by the uncertainties

Table 4. Average tritium concentrations in food and the dose equivalents

Location	1967		1968	
	Concentration (nCi/liter)	Dose equivalent (mrem)	Concentration (nCi/liter)	Dose equivalent (mrem)
Fla: Tampa	0.38	0.06	0.14	0.02
La: New Orleans	.60	.10	.14	.02
Ark: Little Rock	.34	.06	.13	.02
Ky: Louisville	.71	.12	.26	.04
N. Mex: Albuquerque	.48	.08	.24	.04
S. C: Charleston	.69	.12	.26	.04
Miss: Columbia	.40	.07	.18	.03
Tex: Austin	.36	.06	.23	.04
Annual average	0.49	0.08	0.20	0.03

associated with the urine values (*3*) and the fact that an additional source of tritium intake, i.e., drinking water, is not considered if the DE calculation is based on food intake.

The above calculations are valid for standard man with a body weight of 70 kg and a hydrogen body content of 7,000 grams. The DE to children who actually consumed this food depends upon the body weight and the hydrogen content of the body. An estimate of DE of children may be carried out by the method described elsewhere (*3*).

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